Evidence for moving breathers in a layered crystal insulator at 300K

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Abstract

We report the ejection of atoms at a crystal surface caused by energetic breathers which have travelled more than 10^7 unit cells in atomic chain directions. The breathers were created by bombardment of a crystal face with heavy ions. This effect was observed at 300K in the layered crystal muscovite, which has linear chains of atoms for which the surrounding lattice has C_2 symmetry. The experimental techniques described could be used to study breathers in other materials and configurations.

The transport of energy through crystals has been studied extensively in metals, but less so in insulators, in which the usually strong electron-phonon coupling pathway for dispersion of energy is absent. This absence allows long-range transport phenomena involving large amplitudes of motion of atoms about their equilibriums positions to be studied. Large amplitudes of motion are created when a crystal is irradiated by bombardment with energetic ions. Under irradiation some material is sputtered backwards from the surface but most of the incident energy is deposited in the body of the crystal. If the incident energy is sufficiently high, a plasma is created and leads to the development of atomic cascades in which atoms are displaced permanently from their equilibrium positions in the lattice. This stage in the coupling of energy to the lattice can be studied by molecular dynamics (MD), which shows the emergence and distribution of permanently displaced atoms [1, 2, 3]. A notable feature of such cascades is the creation of focused collision sequences, often terminating in an interstitial [4, 5]. These sequences are unstable against thermal motion because they depend on the alignment of atoms [6]. Typically, at ambient temperatures, such sequences are limited to a length of about ten unit cells. Eventually, as the incident energy is dispersed, atomic displacements are no longer possible. It is at this stage, when the available kinetic energy still far exceeds that of phonons, that on-site potentials and long range co-operative interactions between atoms

can influence the subsequent dispersal of energy in the lattice by the creation of self-focusing breathers [7, 8]. Here we present evidence for the existence of energetic, mobile, highly localised, lattice excitations that propagate great distances in atomic-chain directions in crystals of muscovite, an insulating solid with a layered crystal structure. Specifically, when a crystal of muscovite was bombarded at a given point, atoms were ejected from remote points on another face of the crystal, lying in atomic chain directions and at more than 10^7 unit cells distance from the site of bombardment. This effect was predicted from studies of inelastic scattering of breathers using analogue mechanical-magnetic arrays and has been confirmed by numerical simulations. As this experiment was performed with the crystal at around 300K, it demonstrates the stability of these breathers against thermal motions of the lattice.

The evolution of an impulse to a single particle in a discrete particle system consisting of a chain of particles with nonlinear nearest neighbour interactions has been studied extensively and leads to solitons [9]. However, in two-dimensional arrays of such chains these solitons are unstable, limiting flight paths to about 100 unit cells [10] In real crystals the evolution of an impulse is strongly influenced by the surrounding lattice, which introduces on-site potentials. Both mechanical analogues and numerical methods have been used to study how an impulse to a single atom in a chain evolves under these conditions, in particular, for the muscovite lattice. On a time scale of a few picoseconds, it evolves via anharmonic oscillations into a highly localised, mobile, excitation in which adjacent atoms in a chain move in nearly anti-phase motion within an envelope extending over a small ($\sim$10) number of atoms. The envelope also extends in a lateral direction, but most of the energy resides on a single chain. These excitations are a type localization called variously an Intrinsic Localised Mode or a breather [8, 11]. First order numerical modelling of the mica lattice showed that these breathers could propagate up to about 10^4 unit cells before becoming unstable [12].

The requirement that atoms do not become interstitial sets an upper limit on the energy of individual atoms in a breather of $\sim$50 eV. This estimate is derived from plots of potential energy versus displacement, calculated for the muscovite lattice by molecular dynamic methods. A mechanical analogue using magnetic dipoles was constructed, which mimicked the potential energy versus displacement plot. This analogue showed the evolution of an initial impulse into a moving breather, its behaviour on meeting defects in the chain, such as different masses or a vacancy, and reflection at the end of a chain. These studies indicated that breathers could have energies from a few eV up to $\sim$100 eV. Further, it showed that breathers with more than $\sim$10 eV could suffer inelastic scattering at the end of a chain and eject the last atom. This process is illustrated in Fig. 1. The prediction that energetic breathers could eject atoms is testable and was the rationale for the experiment reported here.

The experiment is shown schematically in Fig. 2. It consisted of an electrically insulated rotatable specimen holder placed at the centre of a vacuum chamber and positioned opposite an exit port to which a Photonis X919AL electron channel-plate multiplier [ECM] was attached. This exit port was covered

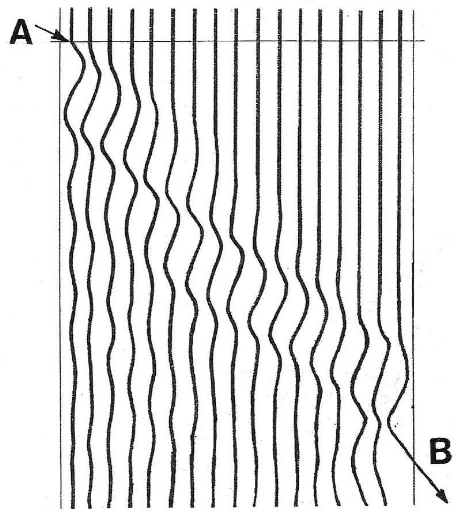

Figure 1: Analogue plot of the longitudinal motion of particles lying in a chain when the first particle is given an impulse at point A. The impulse evolves into a breather that propagates along the chain. Inelastic scattering of the breather when it reaches the end of the chain causes the last particle to be ejected over a potential energy barrier, as shown at point B.

by a fine wire grid to help shape the electric field between the specimen and the ECM when a positive potential of several kV was applied to the specimen holder. A small crystal of muscovite of $\sim$1mm thickness and $\sim$7mm across the (001)-face with good natural edge faces was mounted on the specimen holder with the axis of rotation of the holder normal to the (001)-face. One edge of the crystal was bombarded with α-particles of $\sim$5Mev from an Am^{241} source A located at the bottom of a collimating tube, which also shielded the ECM from the α-particles. This tube was positioned to irradiate one edge of the crystal at near grazing incidence and approximately normal to the (001)-face. A positive potential was applied to the crystal holder to create an electric field approximately normal to the edges. Atoms ejected from the crystal edges and ionized by electrons moving in this electric field were accelerated towards the ECM for detection. The count rate of the ECM was measured as the crystal holder was rotated. The result is shown in Fig. 3.

The peak labelled T is caused by a source of alpha particles mounted on the target holder but screened from the crystal for test and calibration purposes

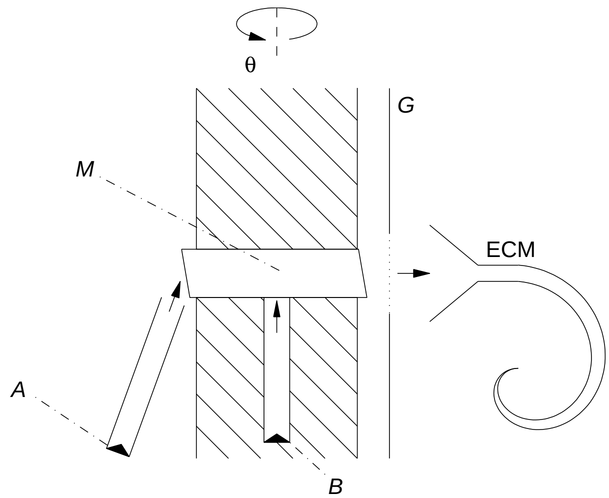

Figure 2: Schematic diagram of the experiment. The crystal of muscovite M was clamped between metal plates pressing on the (001)-faces. In the first arrangement A, the collimated beam of α-particles bombarded one edge. Atoms ejected from the opposite edge, after ionization, passed through the grid G and were detected by the electron channel multiplier ECM. In the second arrangement B, the source is placed below the crystal.

of the detection system. The several peaks labelled S are due to sputtering and back scattering from the irradiated face. Here we are mainly interested in the peak labelled E which is due to atoms ejected from the rear face by inelastic scattering of breathers. The angular position of this peak is consistent with ejection from chains lying in the principal (010) crystal direction, after the breathers had travelled a distance of 10^7 unit cells.

To eliminate the possibility of spurious secondary particles arising from the irradiated front face and then multiply scattered via the chamber to the back face, the reference source was removed and the source used to irradiate the crystal was placed below the crystal in a holder that collimated the alpha-beam, as indicated in Fig. 2 as source B. The source holder could be rotated to bring the source under a hole in the base of the target holder, so that the flux of alphas could be controlled. It was found that the ECM count rate for the peak E was proportional to the alpha flux, as expected.

The absence of a signal from the ECM at zero applied potential showed that the peak E was not due to α-particles channelling through the crystal; the expected range of the alphas in the (001) plane from the source A was less than 10 microns. It was found that the flux of ejected particles decreased slowly in time, with other variables held constant, consistent with a progressive depletion of atoms that could be ejected by inelastic scattering of the breathers. The

decay was approximately exponential in time, with a decay constant of $\sim$4 hours. Possible alternative sources of the ions detected by the ECM were examined. The drop to background count rate either side of the peak E showed that the ions were not arising from out-gassing of the crystal. Also, when a crystal with a freshly cleaved edge was used, it gave similar results to that for a natural edge, implying that the peak E was not arising from surface contamination. This demonstration that breathers can propagate great distances also shows that they can survive the inevitable point defects that occur in real crystals as well as remaining stable in the presence of considerable thermal motion at $\sim$300K. Having remained stable whilst propagating more than 10^7 unit cells, there is no a priori reason to suppose that such breathers could not travel much further in large nearly perfect crystals.

We remark that these results constitute a useful investigative tool for studying the formation and properties of breathers in a wide variety of materials and material configurations. We note also that, although breathers in metals may not have such long path lengths, they may still be important in various effects which are still poorly understood using conventional theories. We conclude by speculating on two possible developments in this area.

Although these results relate to layered crystals, there is evidence that breathers can occur in non-layered crystals, but with shorter path lengths of order microns. This was reported in connection with radiation damage studies in silicon [13] and in diffusion of interstitial ions in austenitic stainless steel [14]. It has also been shown that the lattice conditions required for the existence of breathers, namely linear chains of atoms for which the surrounding lattice has C_2 symmetry, also occur in typical high temperature superconductors [15, 16]. The demonstrated stability of breathers against thermal motion and their ubiquitous occurrence, leads us to predict that reducing breather-breather scattering in the conductive sheets of these superconductors, by separating the sheets into narrow parallel strips, should increase the T_c beyond the present values. This could form a test of the supposition that breathers are involved in pair formation in these layered materials.

It has also been reported that deuterium fusion reactions can be increased by implanting deuterium in a metal [17]. This effect has been explained in part as due to electron screening. Due to the ubiquitous nature of breathers, they will be produced copiously in such experiments. During the evolution of a breather from the initial impact, it is likely that several close encounters occur for each $d-d$ pair, which should contribute to an increased fusion rate. This may be an alternative or an enhancement mechanism for this effect.

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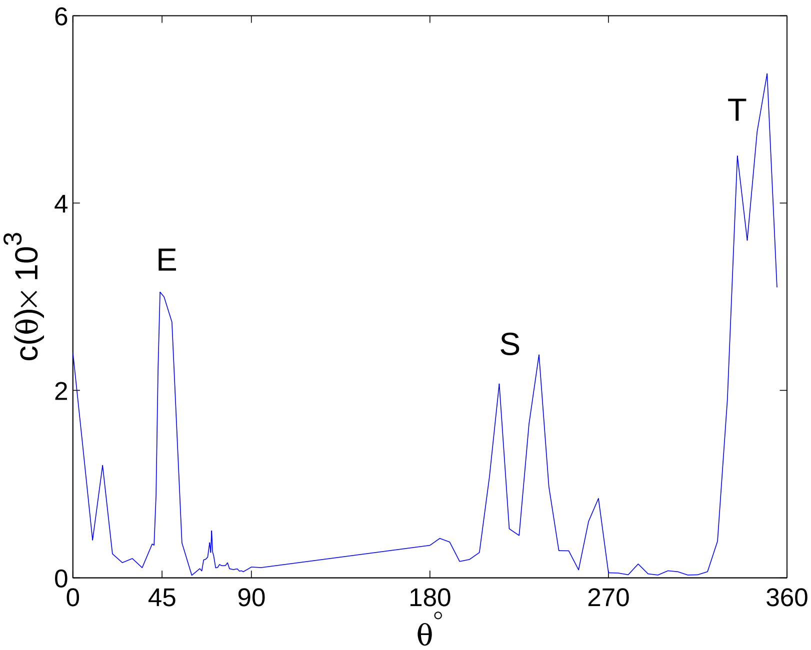

Figure 3: Plot of the angular dependence of the ECM count rate. The peak labelled T arises from a source of alpha particles used for test purposes. The several peaks S arise from both back scattering and sputtering from the front face, when it is irradiated with alpha particles. Breathers are created during the irradiation and propagate in the [010] chain direction. On reaching the rear face of the crystal, after travelling more than 10^7 unit cells through the lattice, those breathers with sufficient energy eject atoms giving rise to the peak labelled E. As the crystal was held at about 300°K, this result demonstrates the stability of breathers against thermal motion, in contrast to the short range of classical collision cascades.